

Thus, the present invention provides a process for producing lithium sulphite, the process comprising introducing sulphurous acid into a reaction vessel and reacting it with lithium carbonate under an inert atmosphere to form an aqueous solution and then evaporating the solution to recover the lithium sulphite. As described in the above 2002 paper, the lithium sulphite may then be used as a precursor to a complexing agent and may be mixed in with a suitable high surface area, N containing pyrolysed polymer in order to improve its properties as an electrode material, especially for supercapacitors. As previously explained, the polymer once combined with lithium sulphite, Li_2SO_3 , and incorporated into an electrode is of the nature of a precursor electrode material, which would normally require an electrochemical charging or formation step to generate N:sulphur trioxide SO_3 complexes at the nitrogen sites.

In a second aspect of the present invention, there is provided a process for producing an electrode composition or material, comprising conducting the process as described above with steps a) to c), wherein between step b) and step c) a nitrogen-carbon structure suitable for use as an electrode material is added to the solution and step c) is then conducted to yield an intimate mixture of $\text{Li}_2\text{SO}_3(\text{s})$ and the nitrogen-carbon structure.

By incorporating the nitrogen-carbon structure into the lithium sulphite synthesis so that lithium sulphite is intimately precipitated in situ around the nitrogen-carbon structure, it is possible to achieve a more homogenised precursor electrode material, and hence one with lower resistance and a better material utilisation. By adapting the lithium sulphite synthesis, the subsequent electrode fabrication process is, of course, also simplified.

By the use of the term "nitrogen-carbon structure suitable for use as an electrode material", we mean, in this context, any carbon based structure which has the requisite nitrogen functionality to form SO_3 complexes and that is suitable for use as an electrode, especially supercapacitor electrodes. Many carbon based structures are known for use as supercapacitor electrodes. As with all electrodes they need to only contain components that do not react unfavourably with the electrolyte, and in the context of supercapacitors they need to have a high surface area, and an open porous structure that will permit ion migration and a robust structure that can withstand the effects of cycling. Supercapacitor electrode materials may typically be formed from activated carbons, or from polymers, for example, electrically conducting polymers (ECP's), or from carbon nanostructures or composites (e.g. ECP coated carbon nanotubes where the nanotubes absorb the mechanical stress) or from thermally restructured polymers which will have increased strength, conjugation and surface area. Especially preferred are any thermally restructured polymers comprised of conjugated arrays with imine functionality (i.e. containing a carbon-nitrogen double bond), whereby the tertiary nitrogen sites can act as hosts to subsequent SO_3 complex formation. Ideally, the thermally restructured polymers are cyclised carbon structures with active pyridinic N-6 (6 membered ring) N sites. For example, the polymer may be formed by the thermal cyclization of nitrile containing polymers such as, for example, acrylonitrile or copolymers thereof, such as fumaronitrile or maleonitrile. When thermally restructured at temperatures of up to 600°C . in an anaerobic environment, conjugated, cyclised arrays of imine functionality are incorporated into such material.

This second aspect of the invention therefore provides a single process step for providing a precursor electrode material that may then be transformed by an electrochemical charging or formation step to generate N:sulphur trioxide

SO_3 complexes at the nitrogen sites, which can then give rise to the increased capacitance (due to pseudo-capacitance).

By way of illustration, FIG. 5 below shows cyclic voltammograms for various TR-PAN electrodes recorded in three-electrode measurement mode (i.e. using a Swagelok test cell) at a scan rate of 10 mV/s. A TR-PAN electrode modified with Li_2SO_3 to form complexed TR-PAN (prepared by an electrochemical formation step) is compared with an unmodified TR-PAN electrode and a commercial electric double layer electrode. The peaks in the electrodes containing Li_2SO_3 are due to "doping and de-doping" of the N— SO_3 complex (or, more correctly, migration of charge from solution species in the electrolyte on and off the complex sites) formed during the formation step. These peaks are noticeably absent in the baseline TR-PAN electrode and commercial EDLC activated carbon benchmark (i.e. a typical EDLC box-like trace). The specific capacitance at 3.5 V (vs Li/Li+) for the TR-PAN, commercial activated carbon and modified TR-PAN respectively, is 110 F/g, 100 F/g and 400 F/g. Results indicate that modifying by use of Li_2SO_3 as a complexing agent has enhanced the capacitance by 4 times.

In a third aspect of the present invention, there is provided a process for forming an electrode material comprising a complexing step of causing lithium sulphite to form SO_3 complexes at active N sites of a nitrogen-carbon structure, in the presence of a selected amount of a sink that absorbs the liberated lithium, so as to form the N: SO_3 complexed electrode material.

The deliberate use of a sink to absorb the lithium ions is novel and provides a superior electrode material from which supercapacitors with very high energy density (capacity) at high power can be produced. Rather than merely allowing the liberated lithium to be absorbed by a traditional counter electrode (e.g. carbon cloth), where it could still be electrochemically active during cycling, it is preferable deliberately to provide a selected amount of a sink capable of efficiently removing the lithium, preferably in an irreversible fashion. The sink also ensures that the complexing reaction is driven to completion.

The nitrogen-carbon structure is as hereinbefore defined. Especially preferred are any thermally restructured polymers comprised of conjugated arrays with imine functionality (i.e. containing a carbon-nitrogen double bond), whereby the tertiary nitrogen sites can act as hosts to subsequent SO_3 complex formation. Ideally, the thermally restructured polymers are cyclised carbon structures with active pyridinic N-6 (6 membered ring) sites. For example, the polymer may be formed by the thermal cyclization of nitrile containing polymers such as, for example, acrylonitrile or copolymers thereof, such as fumaronitrile or maleonitrile. Due to the low cost and availability of PAN, the use of thermally restructured polyacrylonitrile (TR-PAN) is highly preferred.

The nitrogen-carbon structure will contain a variety of different N sites, but not all of these will be active N sites. By "active nitrogen sites", we mean those N sites capable of complexing with SO_3 . It is highly desirable to determine the amount of active N sites in the structure i.e. ones with the functionality and accessibility to be capable of complexing and hence providing pseudo-capacitance. For example, in TR-PAN of the N-6, N-5 and N-Q sites, only the N-6 sites will be active sites. Hence, the process will usually include an earlier step of calculating the amount of active N sites in the nitrogen-carbon structure.

To avoid excess lithium sulphite in the electrode material, the lithium sulphite should be used roughly in an equimolar ratio with the amount of active N sites in the nitrogen-carbon structure (+/-5%). Since the function of the sink is to absorb